Indian J. Phys. **75A** (4), 443-447 (2001)

IJP A
— an international journal

Photoplastic effect in coloured alkali halide crystals

B P Chandra*, R K Pandey and Rashmi Jain

Department of Post Graduate Studies and Research in Physics & Electronics,
Rani Durgavati University, Jabalpur, Madhya Pradesh-482 001, India

Abstract Considering the basic process of photoplastic effect (PPE), an expression is derived for PPE, which reads as

$$\Delta\sigma = \frac{D\delta_2 \varepsilon r_i^0 \gamma_2 A n_{F^0} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_\varepsilon\varepsilon)}\left[\exp(-\alpha_\varepsilon\varepsilon)\exp(-\delta\varepsilon/\dot{\varepsilon})\right]\left\{1-\exp\left[-\delta_0(t-t_c)\right]\right\}$$

The proposed theory is able to explain temporal, deformation, strain- rate, spectral, light intensity, coloration, dimension and thermal characteristics of PPE. PPE can be used to determine the life time of the dislocation captured F^0-centres in crystals and the pinning time of dislocations. It is shown that the lifetime of dislocation-captured F^0-centres is longer as compared to the lifetime of normal F^0-centres, *i.e.*, the dislocation non captured F^0-centres present in the bulk.

Keywords Photoplastic effect, dislocations, alkali halides

PACS Nos. 78.55.Fv, 61.72.Hh.

1. Introduction

Photoplastic effect (PPE) is a phenomenon in which the flow stress of materials is appreciably affected by illumination of light with a wavelength around the intensive absorption. The effect is almost reversible and it can be either negative (softening of the crystal under illumination) as in GaAs or positive (hardening) as in most II-VI compounds [1,2]. The phenomenon of PPE is also known as photo-mechanical effect.

PPE has been observed in alkali halides, semiconductors and other materials. In alkali halides, PPE has been known to be related to the interaction between colour centers and dislocations. Crystals which contained no colour centres did not exhibit the PPE at any temperature. When the sample is illuminated by a light with certain wavelength during the plastic deformation, the increase or decrease in flow stress occurs because the photo-ionized electrons from F-centres influence the motion of dislocations. On the basis of these observations, PPE in alkali halides has been explained by several investigators [3-7], some of which have proposed an interaction of electrons with charged dislocations. However, it has not yet been made clear how the photo-ionized electrons influence the motion of the charged dislocation.

Corresponding Author

The present paper reports a theory of PPE, which is based on the interaction between moving dislocations and photo-obstacle produced during the photoionization process.

2. Theory

From the several experiments, it has been confirmed that photoplastic effect is closely related to the excitation of F-centres. Typical photochemical process of F-centres during F-bleaching at around room temperature may be expressed as

$$2F \xrightarrow{h\nu} F' + F^+ ,$$

where F' is the centre having two electrons in a negative ion vacancy and F^+ is the negative ion vacancy without trapped electrons.

Nadeau [3] has shown that F^+ centre can be eliminated as hardening centre at around room temperature. Thus, the interaction between the positively charged dislocations and F'-centre may be responsible for the PPE.

On illuminating F-centres, equilibrium state of $F \rightleftharpoons F'$ is accomplished in a few seconds. Dislocations are assumed to have constant equilibrium charges (ρ) per unit length and they can interact with F"- centres only in a cylindrical region of radius R around dislocations with appropriate mean strength ($\bar{f}$).

Dislocation densities are expected to increase with the increase of the strain (ε). In this case, the interaction could be expected to increase in the crystal with plastic deformation. Ermakov *et al* [8] have shown that the photo-obstacle centres, responsible for PPE in alkali halide crystals are F-centres.

When a coloured alkali halide crystals is plastically deformed, the movement of dislocations takes place. The moving dislocations may capture electrons from the colour centres and may subsequently transport the captured electrons to hole centres, deep traps and other compatible traps in the crystal. As a matter of the fact, deformation bleaching of the coloration in alkali halide crystals may take place. Suppose a crystal contains N_d dislocations of unit length per unit volume. When N_d dislocations will move through a distance dx, then the area swept out by the dislocations will be $N_d dx$. The deformation bleaching in coloured Alkali halide crystals takes place due to the transfer of electrons from F-centres to the dislocation band and their subsequent recombination with other centres. Near an edge dislocation, some of the F-centres lie in the expansion region and some of the F-centres lie in the compression region. In the expansion region, the energy gap between ground state of F-centre level and dislocation band which lies just above the ground state of F-centre level [9, 10] decreases due to the decrease in the local density of the crystal, however, the energy gap between the ground state of F-centre level and dislocation band increases in the compression region of the dislocation due to the increase in the local density of the crystal. Since, there is a greater probability of the transfer of electrons from the F-centre lying in the expansion region rather than from the compression region of the edge dislocations, the interaction volume may be taken only along the expansion region of the crystals. Thus, the volume in which N_d dislocations interact while moving through a distance dx may be given by $N_d dx\ r_F$, where r_F is the distance up to which a dislocation can interact with the colour centres.

If n_F is the number of F-centres in unit volume, then the number of colour centres interacting with the dislocations will be $N_d n_F r_F dx$. If a dislocation moves the distance dx in time dt, then the rate of interaction g_i of the number of F-centres with dislocations may be given by

$$g_i = N_d n_F r_F \frac{dx}{dt} = N_d n_F r_F v_d ,$$

or
$$g_i = \frac{c}{b} n_F l_F , \tag{1}$$

where v_d is the average velocity of dislocations, $\dot{\varepsilon} = N_d b v_d$ is the strain rate, and b is the Burgers vector.

It has been shown in our previous paper [11] that the density n_F of the F-centres in coloured alkali halide crystals decreases with post irradiation deformation of the crystals, and the dependence of n_F on the strain ε may be expressed as

$$n_F = n_{F^0} \exp(-\alpha \varepsilon). \tag{2}$$

In the PPE experiment, the sample is excited by exposing it to light at $t = t_c$.

If γ_1 and γ_2 are the rate constants for the transformation of exited F- centres into normal F- centres, and for the formation of F'-centres from the excited F- centres, respectively, then we may write the following rate equation

$$\frac{dn_e}{dt} = g_e - \gamma_1 n_e - \gamma_2 n_e , \tag{3}$$

where n_e is the number of excited F-centres at any time t. Integrating eq. (3) and taking $n_e = 0$, at $t = t_c$, we get

$$n_e = \frac{g_e}{(\gamma_1 + \gamma_2)} \left\{1 - \exp\left[-(\gamma_1 + \gamma_2)(t - t_c)\right]\right\}. \tag{4}$$

Thus, the rate of generation of F'-centres may be written as

$$G_{F'} = \gamma_2 n_e = \frac{\gamma_2 g_e}{(\gamma_1 + \gamma_2)} \left\{1 - \exp\left[-(\gamma_1 + \gamma_2)(t - t_c)\right]\right\}. \tag{5}$$

As $1/(\gamma_1 + \gamma_2)$, is the lifetime of excited F-centres and it is very short, $(\gamma_1 + \gamma_2)$ is very high and in equilibrium, the rate of generation of F-centres may be written as

$$G_{F'_s} = \frac{\gamma_2 g_e}{(\gamma_1 + \gamma_2)} . \tag{6}$$

If $\tau = 1/\beta$ is the lifetime of normal F'-centres, then we can write

$$\frac{dn_{F'}}{dt} = G_{F'_s} - \beta n_{F'} , \tag{7}$$

where $n_{F'}$ is the number of F'-centres at any time t. Since the $n_{F'}$ will dependent on the rate of generation and rate of recombination of F'-centres, the contribution of F-centre interacting with dislocations is not taken into account to the right hand side of the eq. (7).

Integrating eq. (7) and taking $n_{F'} = 0$ at $t = t_c$ the time at which light is turned on, we get

$$n_{F'} = \frac{G_{F'_s}}{\beta} \left\{1 - \exp\left[-\beta(t - t_c)\right]\right\}. \tag{8}$$

For longer duration of exposure to light, *i.e.* in equilibrium, eq. (8) may be expressed as

$$n_{F'_s} = \frac{G_{F'_s}}{\beta} . \tag{9}$$

If r_i is the radius of interaction of dislocation with the F'-centres produced during exposure of the sample to light, then similarly to that of the F-centres, the rate of generation of interacting F'-centres may be expressed as

$$g'_i = \frac{\dot{\varepsilon}\, r_i \gamma_2 A n_{F^0} l_L}{b\beta(\gamma_1 + \gamma_2)} \exp(-\alpha \varepsilon). \tag{10}$$

If δ_1 is the constant for the transformation of interacting F'-centres into F-centres by their recombination with F^+-centres ($F' + F^+ \rightarrow 2F$) and δ_2 is the rate constant for the capture of F'-centres by the dislocations, then we can write the following equation

$$\frac{dn_t'}{dt} = g_t' - \delta n_t',$$

where $\delta = (\delta_1 + \delta_2)$.

Now substituting the value of g_t' from eq. (10), we get

$$\frac{dn_t'}{dt} = \frac{\dot{\varepsilon} r_t \gamma_2 A n_{F^0} l_L}{b\beta(\gamma_1 + \gamma_2)} \exp(-\alpha\varepsilon) - \delta n_t', \tag{11}$$

where n_t' is the number of interacting F'- centres at any time t and $1/\delta$ is the lifetime of interacting F'-centres or the pinning time of dislocations [12].

If α_0 is the deformation coefficient of the increase in radius of interaction [13] between the dislocation and F'-centres, then we may write

$$r_t = r_t^0(1 + \alpha_0\varepsilon), \tag{12}$$

where r_t^0 is the value of r_t for undeformed crystals.

For $\alpha_0\varepsilon << 1$, eq. (12) may be expressed as

$$r_t = r_t^0 \exp(\alpha_0\varepsilon) \tag{13}$$

From eqs. (11) and (13), we get

$$\frac{dn_t'}{dt} = \frac{\dot{\varepsilon} r_t^0 \gamma_2 A n_{F^0} l_L}{b\beta(\gamma_1 + \gamma_2)} \exp(-\alpha_e \dot{\varepsilon} t) - \delta n_t', \tag{14}$$

where $\alpha_e = (\alpha - \alpha_0)$.

Integrating eq. (14) and taking $n_t' = 0$ at $t = 0$, we get

$$n_t' = \frac{\dot{\varepsilon} r_t^0 \gamma_2 A n_{F^0} l_L}{b\beta(\gamma_1 + \gamma_2)(\delta - \alpha_e\dot{\varepsilon})}\left[\exp(-\alpha_e \dot{\varepsilon} t) - \exp(-\delta t)\right]. \tag{15}$$

Thus, the rate of generation of dislocation-captured F'-centres may be written as

$$g_d = \frac{\delta_2 \dot{\varepsilon} r_t^0 \gamma_2 A n_{F^0} l_L}{b\beta(\gamma_1 + \gamma_2)(\delta - \alpha_e\dot{\varepsilon})}\left[\exp(-\alpha_e\varepsilon) - \exp\left(\frac{-\delta\varepsilon}{\dot{\varepsilon}}\right)\right]. \tag{16}$$

If $\tau_d = 1/\delta_0$ is the lifetime of dislocation-captured F'-centres, then we can write the following rate equation

$$\frac{dn_d}{dt} = g_d - \delta_0 n_d, \tag{17}$$

where n_d is the dislocation captured F'-centres at any time t.

For low stain – rate, g_d may be taken as constant during the measurement of single PPE, which is completed in a short duration. Thus, integrating eq. (17) and taking $n_d = 0$, at $t = t_c$, we get

$$n_d = \frac{g_d}{\delta_0}\left\{1 - \exp\left[-\delta_0(t - t_c)\right]\right\}. \tag{18}$$

From eqs. (16) and (18), we get

$$n_d = \frac{\delta_2 \dot{\varepsilon} r_t^0 \gamma_2 A n_{F^0} l_L}{\delta_0 b\beta(\gamma_1 + \gamma_2)(\delta - \alpha_e\dot{\varepsilon})}\left[\exp(-\alpha_e\varepsilon) - \exp\left(\frac{-\delta\varepsilon}{\dot{\varepsilon}}\right)\right]\left\{1 - \exp\left[-\delta_0(t - t_c)\right]\right\}. \tag{19}$$

It is known that the force caused by a uniform shear stress σ is $F = b\sigma$ per unit length of dislocation [14]. Thus, the photoplastic effect $\Delta\sigma$ may be given by [15]

$$\Delta\sigma = \left(\pi r_t^2 l\right)\frac{w_0}{b} = \frac{\pi r_t^2 \bar{f} n_d}{b} = D n_d, \tag{20}$$

where $D = \left(\pi r_t^2 \bar{f}\right)/b$ is a constant.

Now, substituting the value of n_d from eq. (19), we get

$$\Delta\sigma = \frac{D\delta_2 \dot{\varepsilon} r_t^0 \gamma_2 A n_{F^0} l_L}{\delta_0 b\beta(\gamma_1 + \gamma_2)(\delta - \alpha_e\dot{\varepsilon})}\left[\exp(-\alpha_e\varepsilon) - \exp\left(\frac{-\delta\varepsilon}{\dot{\varepsilon}}\right)\right]\left\{1 - \exp\left[-\delta_0(t - t_c)\right]\right\} \tag{21}$$

or

$$\Delta\sigma = (\Delta\sigma)_s\left\{1 - \exp\left[-\delta_0(t - t_c)\right]\right\}, \tag{22}$$

where $(\Delta\sigma)_s =$

$$\frac{D\delta_2 \dot{\varepsilon} r_t^0 \gamma_2 A n_{F^0} l_L}{\delta_0 b\beta(\gamma_1 + \gamma_2)(\delta - \alpha_e\dot{\varepsilon})}\left[\exp(-\alpha_e\varepsilon) - \exp\left(\frac{-\delta\varepsilon}{\dot{\varepsilon}}\right)\right]. \tag{23}$$

3. Experimental support to the present theory

Figure 1 shows the PPE for NaCl and KCl crystals measured by Inoue *et al* [15] at room temperature. When the sample is illuminated during deformation, rise in the flow stress is

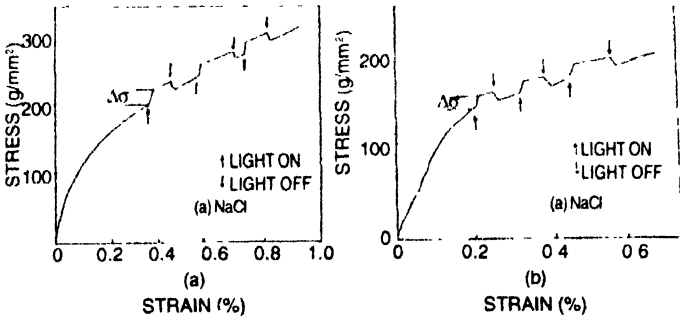

Figure 1. Photoplastic effect for (a) NaCl and (b) KCl measured at room temperature. Turn on and off of illumination are marked by arrows (strain rate 1.2×10^{-4} s^{-1}) (after Inoue *et al* [15])

observed. A region of steady flow state keeps during illumination. When the light is turned off, the flow stress decreases exponenntially to the initial stress level. The result illustrated in Figure 2 indicates the linear increase of $\Delta\sigma$, when the light is turned on [6]. Such linear increase of $\Delta\sigma$, attainment of saturation value and exponential decay are indicated by eq. (22).

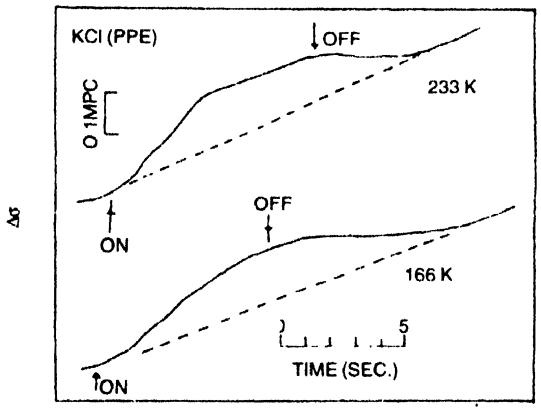

Figure 2. Typical examples of the PPE in coloured KCl (strain rate 5×10^{-5} s^{-1}) (after Hagihara *et al* [61])

The decay time of dislocation captured F^{II}-centres determined from the rise of PPE in KCl crystals with time is determined using eq. (22) and it is found to be 2.5 sec and 2.0 sec for 166 K and 233 K, respectively. The decay time of dislocation-captured F^{II}-centres is determined for NaCl crystals and is found to be 1.6 sec [3]. Figure 3 shows the dependence of $\Delta\sigma$ on the strain level, for KCl crystal, where the experimental data have been taken from the results reported by Inoue *et al* [15] and the theoretical data are based on eq. (23), here $\Delta\sigma_0$ corresponds to the value of $\Delta\sigma$ for zero strain level.

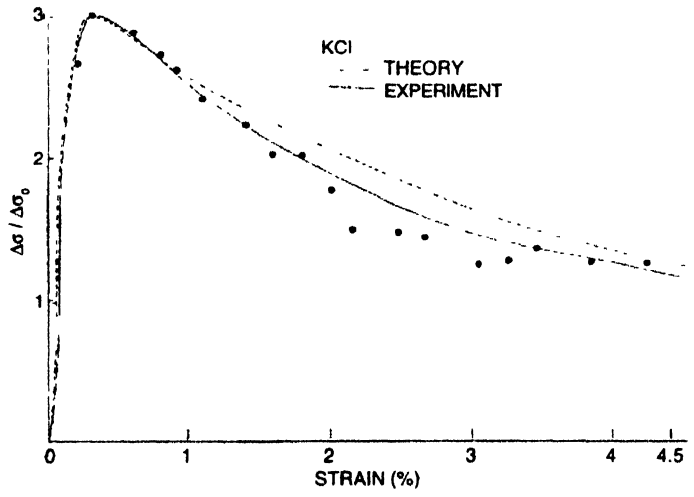

Figure 3. The dependence of the photoplastic effect on strain level for KCl crystals having F-centre concentration $n_F = 4.7 \times 10^{16}$ cm^{-3}. Illumination was done with monochromatic light of 565 nm which lies near the F-peak wavelength. The magnitude of $\Delta\sigma$ is normalized by that of $\Delta\sigma$ at $\varepsilon = 0$, that is, $(\Delta\sigma_0)$ (after Inoue *et al* [15]).

Hagihara *et al* [5] have studied the strain – rate dependence of PPE for coloured KCl crystals at 166 K and it has been found that $\Delta\sigma$ increases linearly with the strain rate. Cabrera and Agullo- Lopertz [7] have studied the dependence of PPE on light intensity for KCl crystal. It is found that, initially, the PPE increases linearly with light intensity and then it tends to attain a saturation value for higher intensity of the light.

Figures 4(a) and (b) shows the dependence of $\Delta\sigma / \Delta\sigma_F$ on the wavelength (or photon energy) of exciting light [15] for NaCl and KCl crystals, respectively, where $\Delta\sigma_F$ is the magnitude for $\Delta\sigma$ given at the same strain level in the case of F-light illumination, 465 nm for NaCl and 565 nm for KCl, respectively. Solid curve indicates the optical absorption spectra near the F - band peak. Both curves roughly resemble the respective F - absorption bands. Such fact is expected from the proposed theory.

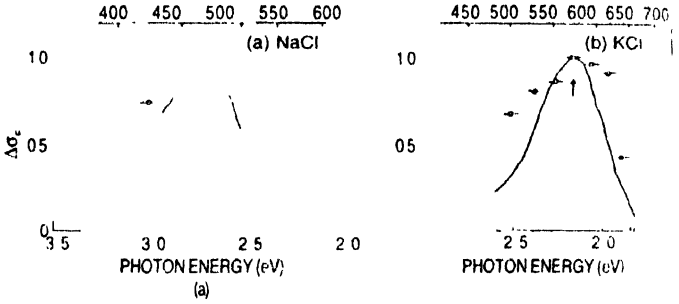

Figure 4. Dependence of the photoplastic effect on wavelength (photon energy) of exciting light for (a) NaCl and (b) KCl crystals. centre concentration (N_c) for NaCl and KCl are 1.0×10^{17} cm^{-3} and 4×10^{16} cm^{-3}, respectively. Solid curves correspond to the optical absorption spectra near the F-band peak (after Inoue *et al* [15])

Eq. (23) indicates that for a given strain the PPE should increase linearly with the initial colour centre concentration n_f. This is practically true for samples having low concentration of colour centres where the effect of non-uniform coloration does not occur [7].

Figure 5 shows the kinetics of the stress decay after light is turned off for additively coloured KCl crystals [3]. The

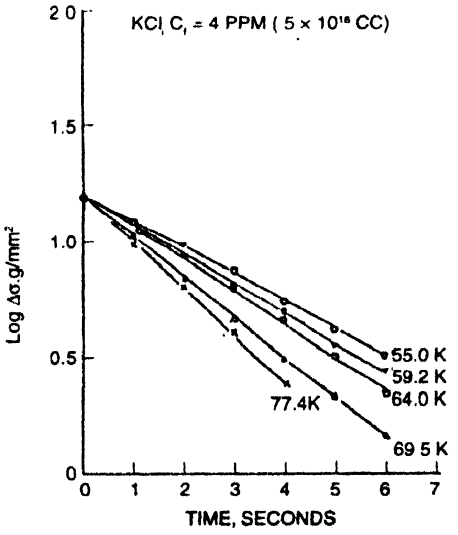

Figure 5. Kinetics of the stress decay after light is turned off. Additively coloured KCl (after Nadeau [3]).

temperature dependence of the rate constant for stress decay follows the Arrhenius plot. This fact shows that the lifetime of dislocation captured F'-centres decreases with increasing temperature of the crystals [3].

4. Conclusions

The important conclusions drawn from the study of PPE in coloured alkali halide crystals are as given below :

(i) Considering the basic process, an expression is derived for PPE.

(ii) The proposed theory is able to explain temporal, deformation, strain-rate, spectral, light intensity, coloration, dimension and thermal characteristics of PPE.

(iii) PPE can be used to determine the lifetime of the dislocation captured F'-centres in crystals and the pinning time of dislocations. It is shown that the lifetime of dislocation-captured F'-centres is longer as compared to the lifetime of normal F'-centres, *i.e.*, the dislocation non-captured F'-centres present in the bulk.

References

[1] B E Madivanyan and M Sh Shikhsaidov *Phys. Stat Sol.* (a) **107** 131 (1988)

[2] Yu A Ossipyan, V F Petrenkov, A V Zaretskii and R W Whitworth *Adv Phys.* **135** 115 (1986)

[3] J S Nadeau *J. Appl Phys.* **35** 669 (1964)

[4] F M Korovkin and Ya M Soifer *Sov Phys Solid State* **18** 1435 (1976)

[5] T Hagihara, Y Hayashiuchi and T Okada *Phys Lett* **108** A 263 (1985)

[6] T Hagihara, Y Hayashiuchi and T Okada *Phys Lett* A **115** 385 (1986)

[7] J M Cabrera and F Agullo-Lopertz *J Appl Phys* **45** 1013 (1974)

[8] G A Ermakov, E V Korovkin and Ya M Soifer *Sov Phys. Solid State* **16** 457 (1974)

[9] D L Dexter *Phys Rev.* **93** 595 (1974)

[10] M I Molotskii *Sov Sci. Rev B Chem* **13** 1 (1989)

[11] B P Chandra, H L Vishwakarma and P K Khare *Phys Stat. Sol.* **B204** 625 (1997)

[12] Y Hayashiuchi, T Hagihara and T Okada *Phys. Lett* A **147** 4 (1990)

[13] R W Whitworth *Adv Phys.* **24** 203 (1975)

[14] C Kittel *Introduction to Solid State Phys* vth edn (New York John Wiley) (1976)

[15] Y Inoue, T Okada and T Hagihara *Technol Rep* Osaka Univ. **29** 33 (1979)